Lightning NO_x Production Estimated from OMI and TROPOMI NO_2 Observations

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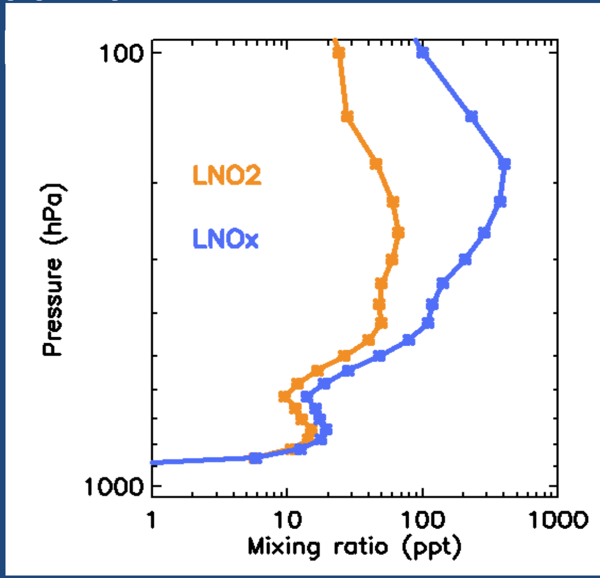
OMI LNO_x Algorithm

$$V_{LNOx}{}^{*} = \frac{S_{total} - Vstra_t \times AMFstrat}{AMF_{LNOx}}$$

- $V_{LNOx}{}^{*}$ = Vertical column amount of NO_x in deep convective clouds from recent lightning and background. Total slant column NO_2 (S_{total}) from spectral fitting (OMI V3.0 Standard Product)

- V_{strat}: Daily zonally-averaged stratospheric NO_2 vertical column derived from OMI Standard Product and stratospheric Air Mass Factor (AMF_{strat})

- AMF_{LNOX} converts the slant column LNO_2 to vertical column LNO_x. AMFs from radiative transfer modeling using assumed profile shapes appropriate for LNO_2 and LNO_x, cloud information, surface albedo. Monthly are from Global Modeling Initiative (GMI) chemical transport model, selected from days with significant NO_2 from lightning.



- Since $V_{LNOx}{}^{*}$ contains contributions of sources other than recent lightning, in-cloud tropospheric background must be considered.

Analysis Methods

- Daily analysis conducted on an array of 1 x 1 degree grid cells for June, July, August 2007 – 2011: Gulf of Mexico (Pickering et al., 2016, *JGR*); three mid-latitude continental regions (Bucsela et al., 2019, in press); three tropical focus areas (Allen et al., 2019, in revision).

- Analysis performed using OMI pixels with large values (≥ 0.97) of Cloud Radiance Fraction (CRF) and with Optical Centroid Pressure (OCP ≤ 500 hPa) sufficiently low to indicate deep convective cloud.

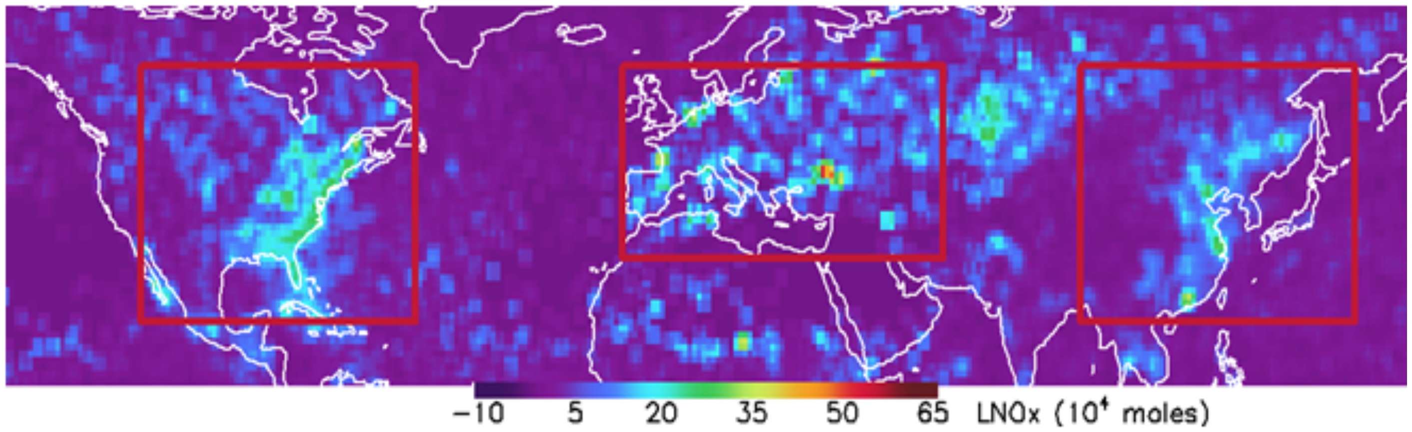
- Analysis conducted only for grid cells with > specified minimum number (3) of OMI pixels meeting CRF and OCP criteria in order to reduce noise.

- Analysis conducted only for grid cells with non-zero flashes in counting window (time period prior to OMI overpass ≤ median residence time for upper tropospheric air in grid cell). Gulf of Mexico: 3 hr; Mid-latitudes: 1 hr; Tropics: tested 1 to 6 hours

- Flash data are from World-Wide Lightning Location Network (WWLLN) corrected for detection efficiency.

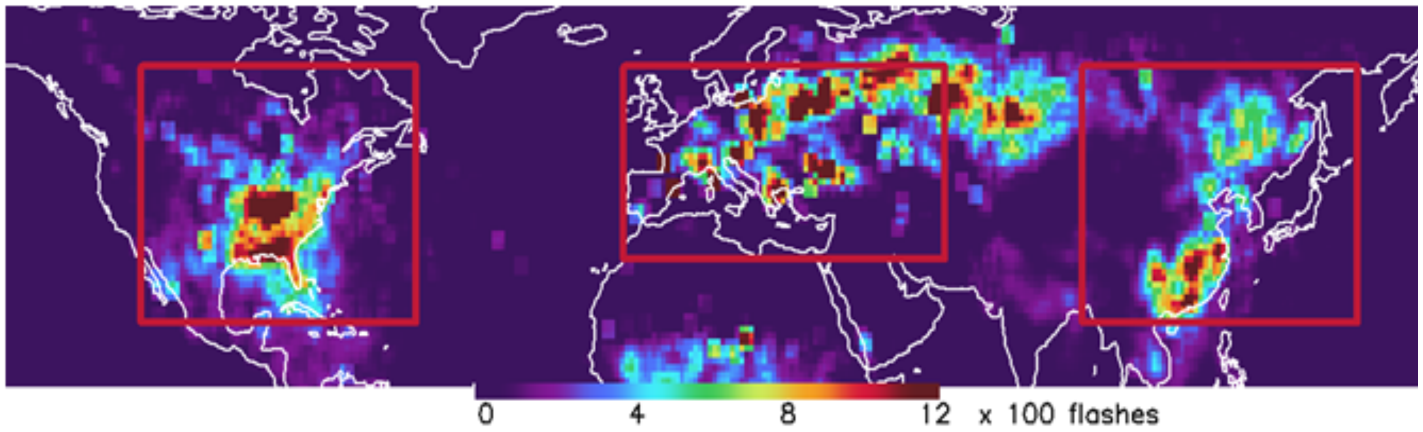
Mid-latitude Analysis

(c)



V_{LNOx}: vertical column of NO_x from recent lightning obtained by subtracting background (assumed to be given by temporal average of $V_{LNOx}{}^*$ over non-flashing days meeting the cloud criteria) from $V_{LNOx}{}^*$ for flashing grid cells

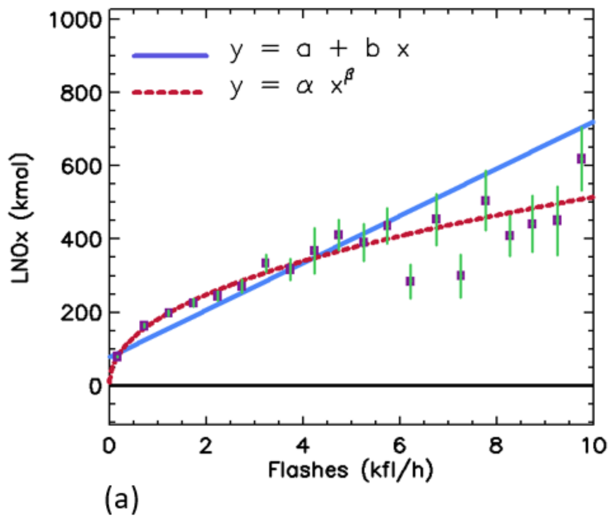
(d)

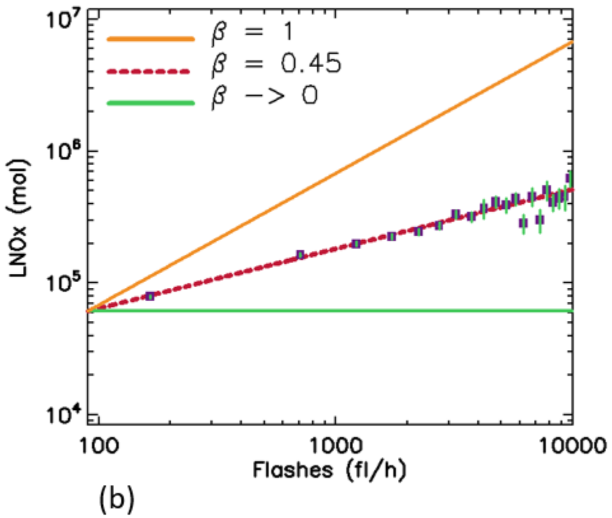


WWLLN flashes in 1-hour window prior to OMI overpass; adjusted for detection efficiency

Red boxes show North American, European, and East Asian regions for which LNOx Production Efficiency (PE, mol/fl) are computed

Regression of Binned Mid-latitude Data





Linear fit: $y = a + bx$

Power law fit: $y = \alpha x^{\beta}$

Power law provides better fit to data than linear regression; suggests lower LNO_x PE at higher flash rates.

Implying lesser LNO_x production per flash with smaller flash extent, the above result is consistent with data from ground-based Lightning Mapping Arrays, which show inverse relation between flash rate and flash extent.

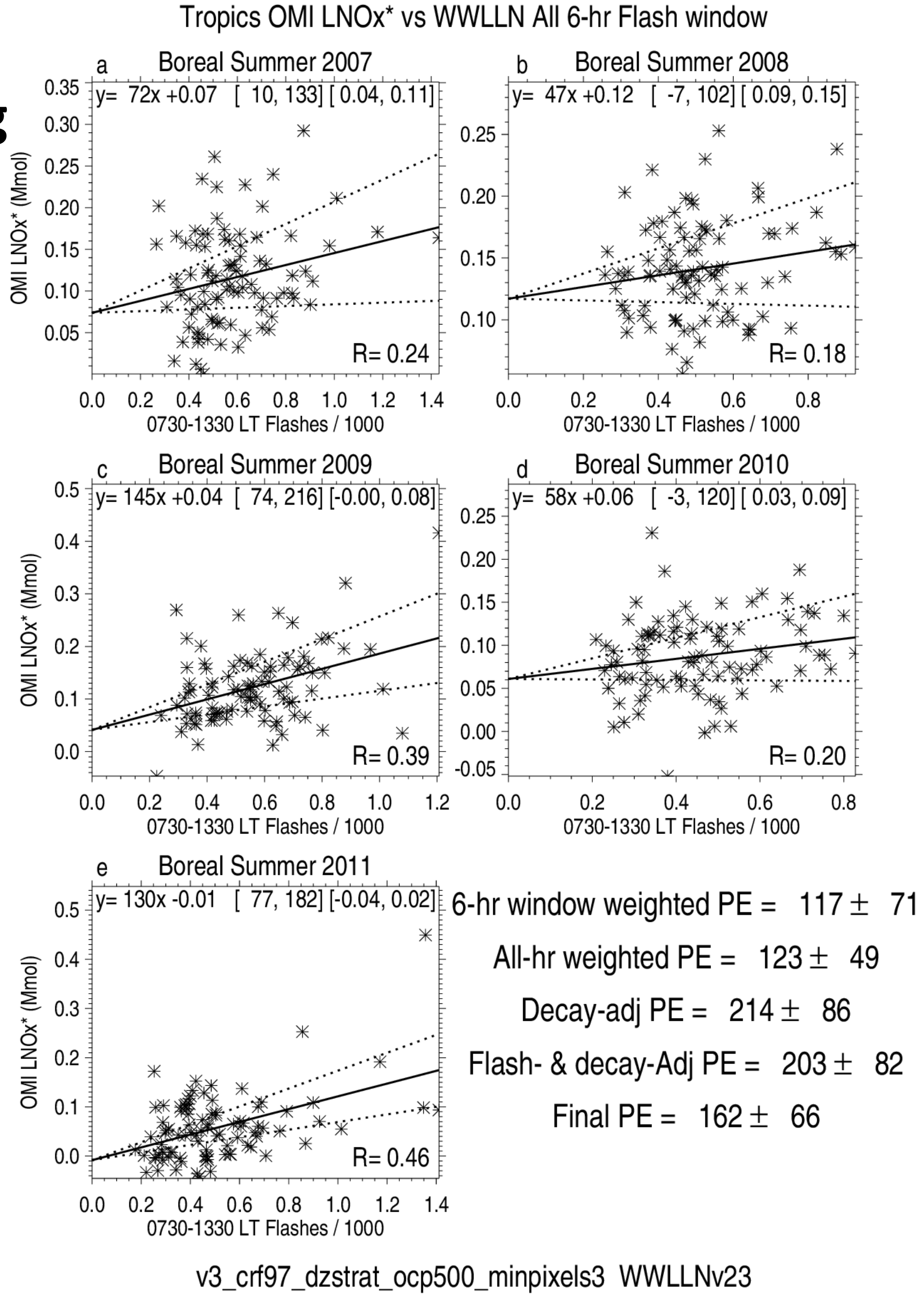
Region	r	β	χ^2_r (linear)	χ^2_r (power)	PE_{avg} (mol/fl)
N. Amer.	0.59	0.42	5.79	1.84	200 ± 110
Europe	0.42	0.39	2.04	1.40	150 ± 90
E. Asia	0.86	0.51	5.77	1.78	160 ± 100
Combined	0.87	0.45	10.1	1.49	180 ± 100

← Computed by summing LNO_x and flashes over the five seasons

Results assume:

- 3-hr NO_x lifetime (Nault et al. 2017)
- WWLLN calibration relative to LIS
- Correction for model NO/NO_x bias (Silvern et al., 2018)

Tropics
June, July, Aug
2007 - 2011



Mean of five individual JJA season analyses

Tropics: 162 ± 66 mol/fl
Tropical Americas: 120 ± 47
Tropical Africa: 107 ± 39
Tropical Pacific: 263 ± 97

Continental: 94 ± 45
Marine: 425 ± 217

← 1, 2, 3, 4, 6-hr values weighted by explained variances

← 3-hr lifetime assumed (Nault et al., 2017)

← Correction for WWLLN bias relative to LIS

← 20% adjustment for AMF low bias due to model NO/NO2 bias (Silvern et al., 2018)

Background assumed to be represented by y-intercept of regression

TROPOMI LNO_x PE Algorithm – Individual Storm Analyses

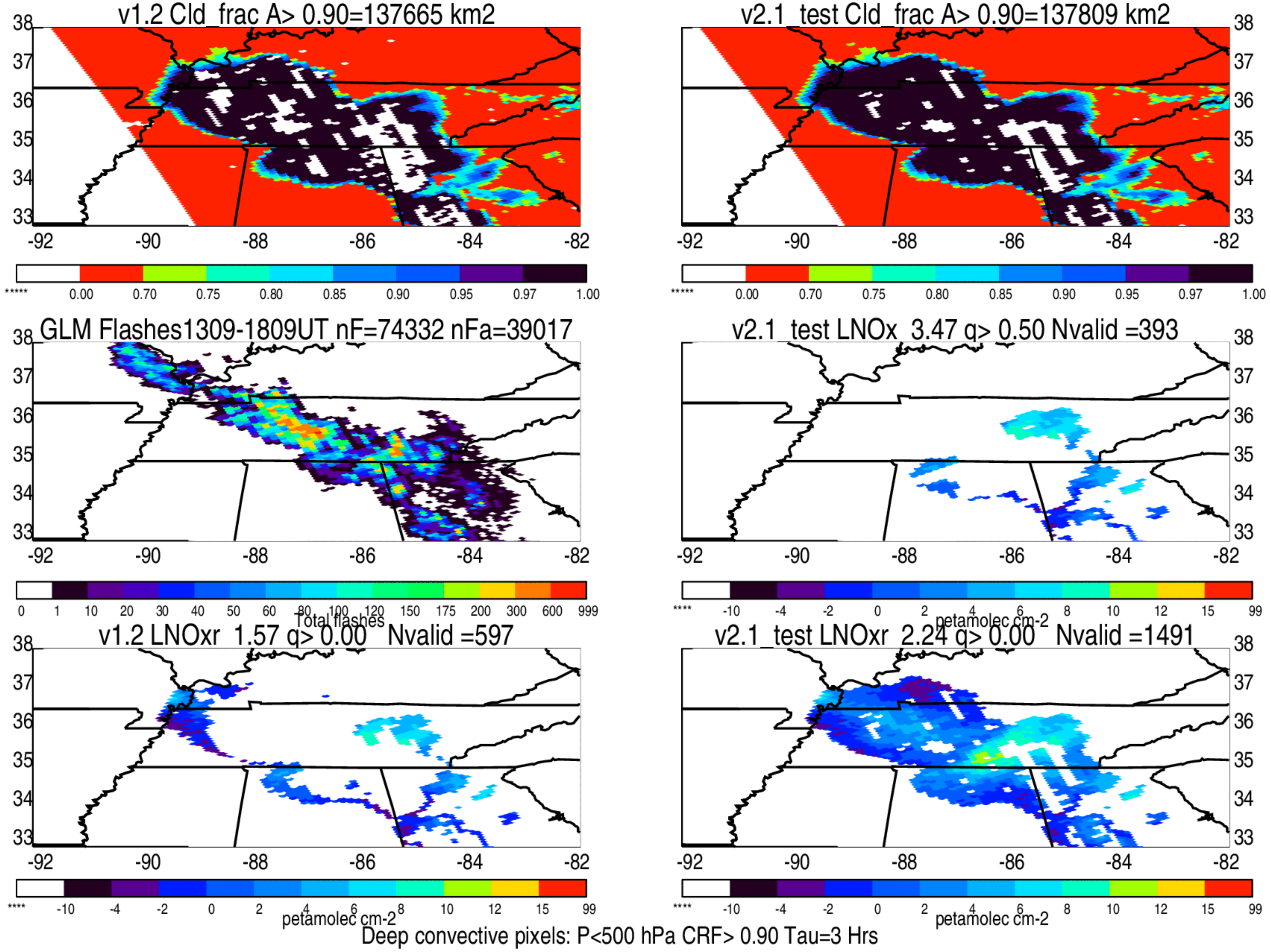
Tropospheric Column Method: $V_{tropLNOxP} = [V_{tropNO2} \times AMF_{trop}] / AMF_{LNOx}$
Tropospheric Residual Method: $V_{tropLNOxP} = [S_{NO2} - V_{stratNO2} \times AMF_{strat}] / AMF_{LNOx}$

S_{NO2} ≡ Total NO_2 slant column for individual deep convective cloud (DCC) pixels within ROI
$V_{tropNO2}$ ≡ Tropospheric NO_2 vertical column for DCC pixels within ROI with a quality flag > 0.50
$V_{stratNO2}$ ≡ Stratospheric vertical column of NO_2 for DCC pixels within ROI
AMF_{strat} ≡ Stratospheric air mass factor for DCC pixels within ROI
AMF_{LNOx} ≡ AMF converting tropospheric slant column of NO_2 to vertical column of LNO_x
0.46 after *Beirle et al*. (2009, ACP)

$$PE = [V_{tropLNOx} \times \Sigma\ AreaP] / [N_A \times \Sigma\ (Flashes \times \exp(-t / \tau))]$$

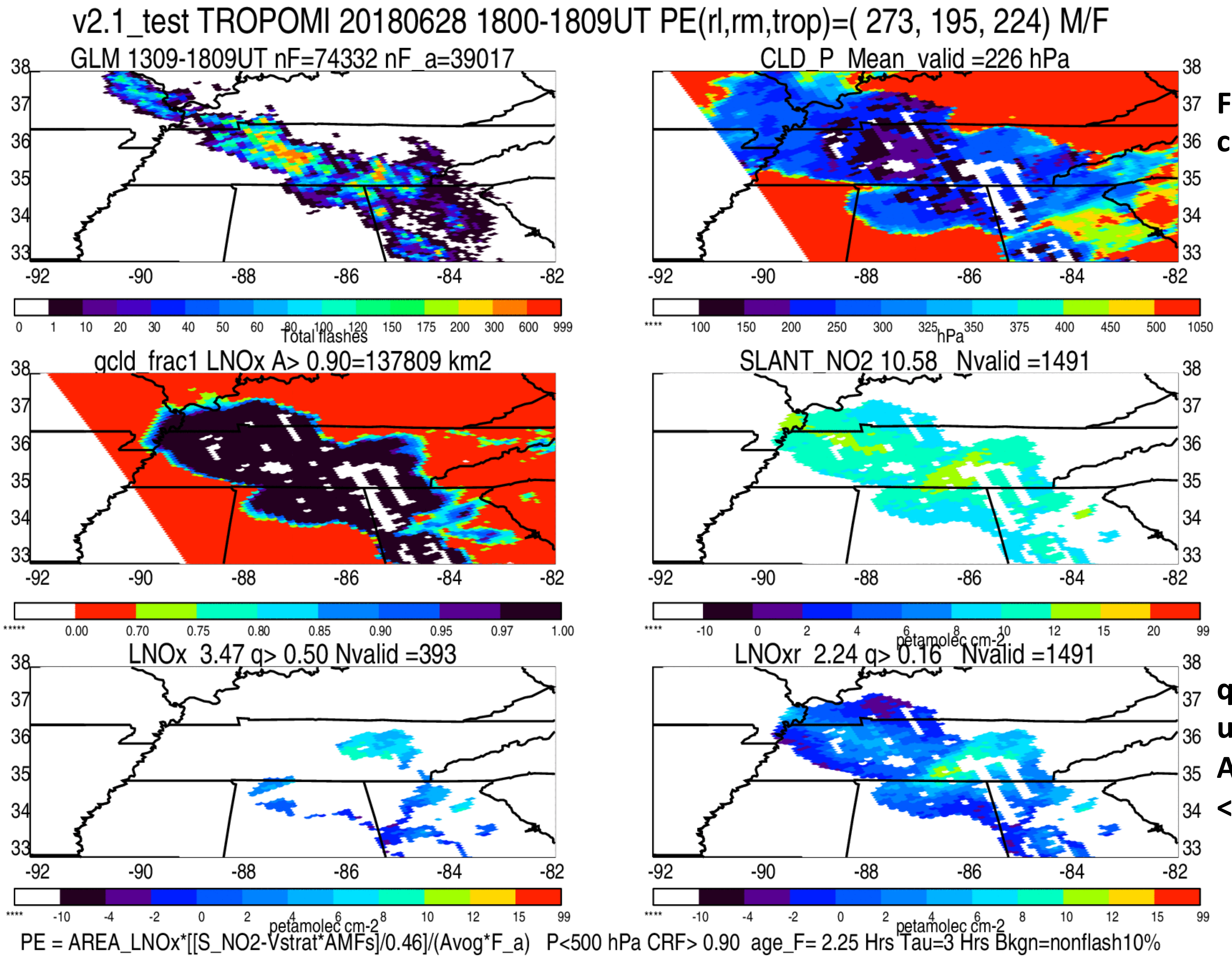
PE ≡ LNO_x Production Efficiency (moles NO_x/flash)
$V_{tropLNOx}$ ≡ Median $V_{tropLNOxP}$ over flashing pixels in ROI minus 10^{th} percentile of $V_{tropLNOxP}$ over non-flashing pixels
AreaP ≡ Area of individual pixels within ROI that satisfy the deep convection constraints (DCC) of cloud fraction > 0.90 and cloud pressure < 500 hPa; includes embedded missing pixels
N_A ≡ Avogadro's Number
Flashes ≡ Number of GOES-16 GLM flashes within ROI during 5-hour period before TROPOMI overpass time
t ≡ Age of individual flashes
τ ≡ Lifetime of NO_2 in near field of convection (assumed to be 3 hours)

Flashes & LNOx: 20180628 Convective System (TROPOMI Overpass time =1800-1809UT)



V1.2 → V1.3
Pixels with AMF < 0.1 AMF_g are now included

V1.3 → V2.1 test
Saturation constraint relaxed



From FRESCO cloud algorithm

Cloud fraction in NO_2 window

q > 0.16 allows use of pixels with AMF_{trop}/AMF_g < 0.1

Summary of LNO_x PE from TROPOMI for Selected US Storms

		From tropospheric NO_2 product V2.1 test		From residual NO_2 product V2.1 test	
YYYYMMDD	Location	N_{valid} pixels ($q>0.5$)	LNO_x PE (mol/F)	N_{valid} pixels ($q>0.16$)	LNO_x PE (mol/F)
20180628	Tennessee	393	224	1491	273
20180628	Iowa	645	247	1901	1011
20180720	Indiana/Ohio	653	64	735	63
20180722	Northern FL	917	121	1178	126
20180726	Off of NC coast	1520	288	1638	270
20180731	Southern MS	1275	145	1547	130
20190608	Southeast US	4013	180	4245	184

Summary

- No significant difference found between LNO_x PE in mid-latitudes and tropics in boreal summer: 180 ± 100 mol/fl mid-latitudes and 162 ± 66 mol/fl tropics

- Mid-latitude LNO_x production is proportional to a power function of lightning flash rate, with exponent between 0.4 and 0.5. Implies that the PE is smaller in storms with more frequent flashes.

- Mean PE largest in low-flash rate tropical Pacific region and smallest in high flash rate tropical Africa

- TROPOMI V2.1 test NO_2 algorithm over deep highly-reflective clouds is best version yet, but operational V1.3 (late March 2019 onward) may also be suitable for LNO_x analysis. Individual storm analyses yield reasonable PE values.